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Effect of Sr substitution on electrical transport and thermoelectric properties of $Y_{1-x}Sr_xCoO_3$ ($0 \le x \le 0.2$) prepared by sol–gel process

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Abstract

The effect of Sr substitution on electrical transport and thermoelectric properties of $Y_{1-x}Sr_xCoO_3$ ($0 \le x \le 0.2$) has been investigated in the temperature range from 20 to 780 K. The $Y_{1-x}Sr_xCoO_3$ samples with different Sr concentrations have been synthesized by using the sol–gel process. The results show that dc electrical resistivity of $Y_{1-x}Sr_xCoO_3$ decreases remarkably with increasing Sr content x, which can be attributed to the reduction in lattice distortion. The Seebeck coefficients S of $Y_{1-x}Sr_xCoO_3$ are all positive, indicating predominant hole-type charge carriers, and they decrease monotonously with increasing Sr content of substitution. Moreover, experiments show that the power factor changes non-monotonously with Sr substitution with a maximum value of 2.69×10^{-5} W/m K^2 at 650 K for the sample $x=0.05$, which is about five times greater than that for $x=0$, indicating that appropriate Sr substitution can effectively improve thermoelectric properties of $Y_{1-x}Sr_xCoO_3$.

Keywords: A. Sol–gel processes; C. Electrical properties; D. Transition metal oxides

1. Introduction

In recent decades thermoelectric materials have renewedly attracted extensive attention due to their great application in clean power generation using the Seebeck effect and refrigeration by the Peltier effect amidst polluted environment and energy crisis. Thermoelectric performance for thermoelectric materials is characterized in terms of a dimensionless figure of merit $ZT=S^2T/(\rho\kappa)$, where S is the Seebeck coefficient or thermopower, ρ is the electrical resistivity, κ is the total thermal conductivity, T is the absolute temperature, and the power factor P is given by the expression $P=S^2/\rho$ [1]. Therefore, a thermoelectric material with good thermoelectric performance should have large thermopower with small electrical resistivity and thermal conductivity.

Recently, metal oxides have been considered as good candidates as thermoelectric materials due to, compared with the traditional materials, such as Bi_2Te_3, $CoSb_3$, $Si_{1-x}Ge_x$ etc, their easy fabrication, low cost, and high thermal and chemical stability at high temperature in air. In 1997, Terasaki et al. [2] reported that the single crystal $NaCo_2O_4$ exhibited a good thermoelectric performance with thermopower $S\sim100$ μV/K and resistivity $\rho\sim200$ μΩ cm at 300 K. One after the other, in 2004, Androulakis and Pantelis Migiakis [3] found a very respectable room-temperature oxide thermoelectric material $La_{0.95}Sr_{0.05}CoO_3$ with the value of figure of merit 0.18. In last several years, our group [4–8], in addition to other authors [9,10], has also considered $YCoO_3$ (a distorted perovskite structure [11,12] with orthorhombic symmetry, space group *Pbnm* Z=4, where six oxygen atoms surround each cobalt ion) as a thermoelectric material and investigated it, because of its large thermopower $S\sim1039$ μV/K [5] and small thermal conductivity $\kappa\sim5.39$ W/m K [5] in the vicinity of room temperature, which are two preconditions for having good thermoelectric performance (i.e. large ZT value). This is already fulfilled for the $YCoO_3$ series. However, it has large electrical resistivity (such as $\rho\sim6.9$ Ω m at 300 K [4]), which is a disadvantage as a thermoelectric material. Our group [4] and Hejtmánek et al. [9,10] have succeeded in decreasing its electrical resistivity by substituting Ca for Y, and we estimated the thermoelectric performance ($ZT\sim0.019$ at 660 K). Our

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investigation and the suggestions given by the researchers for similar system $R_{1-x}Ca_xCoO_3$ (R=Ho, Dy, Gd, Sm, Nd, and Pr) [13–15] indicated that the decrease of lattice distortion degree of $YCoO_3$ by the substitution of large ion radius Ca^{2+} (0.99 Å) for Y^{3+} (0.89 Å) or larger rare-earth ion for small rare-earth ion might be advantageous for decreasing its electrical resistivity and improving its thermopower. Thus, can the substitution of much larger Sr^{2+} (1.12 Å) ($Sr^{2+} > Ca^{2+}$ in ion radius) for Y in $YCoO_3$ can enhance more effectively its thermoelectric performance? Moreover, as far as we know, the electrical transport and thermoelectric properties of the substitution of Sr for Y in $YCoO_3$ have been scarcely investigated except for the investigation of $Y_{1-x}Sr_xCoO_3$ film as a gas sensor by Michel et al. [16]. At present, we report the effect of substitution of Sr for Y in $YCoO_3$ on electrical transport and thermoelectric properties in the temperature range from 20 to 780 K.

2. Experimental details

Strontium doped yttrium oxides were prepared from stoichiometric amounts of strontium, cobalt and yttrium nitrates. $0.02x$ (x=0, 0.01, 0.05, 0.15, and 0.20) mol of $Sr(NO_3)_2$ (99.5%), $0.02(1-x)$ mol of $Co(NO_3)_2 \cdot 6H_2O$ (99.0%), and 0.02 mol of $Y(NO_3)_3 \cdot 6H_2O$ (99.0%) were dissolved in 20 ml of an aqueous solution of 0.02 mol citric acid (99.5%) using de-ionized water. The solutions were continuously stirred for about 1 h, resulting in a homogenization of Sr^{2+}, Co^{2+} and Y^{3+} cations in the mixture solution. Subsequently, the mixtures were slowly heated to 90 °C, and at this temperature they were dried for 12 h forming a brown powder from the mixtures. The brown powders were calcined in air at atmospheric pressure in a muffle furnace at 300 °C for 2 h, forming black powder samples so as to remove organic compounds. Then, these powder samples were ground and subsequently compressed into pellets 13 cm in diameter. Finally, bulk samples were obtained by sintering the green pellets in air in a tube furnace at 900 °C for 120 h, subsequently followed by spontaneous cooling slowly down to room temperature in the furnace.

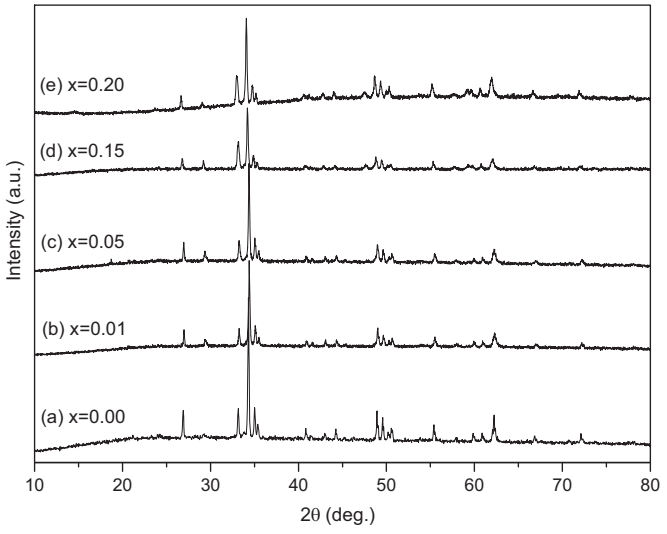

Fig. 1. XRD patterns (CuK_α irradiation) for $Y_{1-x}Sr_xCoO_3$ (x=0, 0.01, 0.05, 0.15, 0.2).

Phase structures and lattice constant for the obtained compounds were characterized by using XRD (Philips-X PERT PRO diffractometer) with Cu K_α irradiation (λ=0.154056 nm) at room temperature. The accurate lattice parameters were determined from the d-value of the XRD peaks using a standard least-square refinement method with a Si standard for calibration. To measure their electrical resistivity, bar-shaped specimens of the size (~)12 mm × (~) 2 mm × (~)1.0 mm were cut from the bulk samples. The electrical resistivity and Seebeck coefficient were measured on a computer-assisted device. DC electrical resistivity was measured in vacuum by a conventional standard four-probe method at the temperatures from 20 to 720 K. Seebeck coefficient was also measured in vacuum in a temperature range from 300 to 780 K. The power factor was calculated from the above-measured S and ρ parameters.

3. Results and discussion

3.1. Phase determination, change of lattice parameters and lattice distortion degree for $Y_{1-x}Sr_xCoO_3$

Fig. 1 presents the XRD patterns of $Y_{1-x}Sr_xCoO_3$ (x=0, 0.01, 0.05, 0.15, and 0.20) with different strontium contents at room temperature. The main diffraction peaks of XRD patterns for synthesized $YCoO_3$ are very consistent with what is indexed based on the standard JCPDS card (No. 88-0425) of $YCoO_3$. We can observe from Fig. 1 that the main peaks of the doped samples $Y_{1-x}Sr_xCoO_3$ ($x \neq 0$) are in agreement with that of the synthesized sample $YCoO_3$, indicating that the substitution of Sr for Y in $YCoO_3$ has taken place and the substituted compounds have formed.

To check the deformation degree of the $YCoO_3$ lattice after Sr substitution, accurate lattice constants a, b and c are measured from the XRD data, as listed in Table 1, whose values are consistent with other literature data [12]. One can

Table 1
The room temperature lattice parameters a, b and c, and unit cell volume V.

x	a (Å)	b (Å)	c (Å)	V (Å^3)
0.00	5.1393($\pm 2.6\times10^{-4}$)	5.4197($\pm 2.7\times10^{-4}$)	7.4588($\pm 3.7\times10^{-4}$)	207.75($\pm$0.01)
0.01	5.1406($\pm 2.6\times10^{-4}$)	5.4182($\pm 2.7\times10^{-4}$)	7.4591($\pm 3.7\times10^{-4}$)	207.76($\pm$0.01)
0.05	5.1437($\pm 2.6\times10^{-4}$)	5.4181($\pm 2.7\times10^{-4}$)	7.4611($\pm 3.7\times10^{-4}$)	207.93($\pm$0.01)
0.15	5.1445($\pm 2.6\times10^{-4}$)	5.4153($\pm 2.7\times10^{-4}$)	7.4602($\pm 3.7\times10^{-4}$)	207.83($\pm$0.01)
0.20	5.1443($\pm 2.6\times10^{-4}$)	5.4204($\pm 2.7\times10^{-4}$)	7.4605($\pm 3.7\times10^{-4}$)	208.03($\pm$0.01)

note that the lattice parameters of $Y_{1-x}Sr_xCoO_3$ change as the substituted Sr content increases. The lattice parameter a, firstly, increases rapidly with the increasing Sr content (such as 5.1393 Å for $x=0$ to 5.1406 Å for $x=0.01$ and 5.1437 Å for $x=0.05$); then the lattice constant a has scarcely obvious change (for instance, 5.1445 Å for $x=0.15$ and 5.1443 Å for $x=0.20$) within the experimental error. Moreover, the lattice constant c also firstly increases from 7.4588 Å for $x=0$ to 7.4591 Å for $x=0.01$ and subsequently to 7.4611 Å for $x=0.05$; then decreases to 7.4602 Å for $x=0.15$ and 7.4605 Å for $x=0.2$. The results, on the other hand, imply that the substitution of Sr for Y in $YCoO_3$ lattice has indeed taken place, and indicate that the Sr substitution leads to the change of distortion degree of $YCoO_3$, which is also reflected in the change of volume of unit cell (calculated from the lattice constants a, b and c) of $Y_{1-x}Sr_xCoO_3$, such as 207.75 $Å^3$ for $x=0$ and 208.03 $Å^3$ for $x=0.2$, as listed in Table 1. From Table 1 we can note that there is no monotonic dependence between composition and unit cell parameters, which can be due to the production of $[Co_{Co}^{\cdot}]$ species (see the following discussion). We also obtain from the calculation of lattice parameters a–c the relation $a < c/\sqrt{2} < b$ for all the samples $Y_{1-x}Sr_xCoO_3$, indicating that lattice structure of these samples belongs to so-called typical O-type structure and the distortion of lattice in $YCoO_3$ is relatively small after Sr substitution [17]. The main source of small distortion is the buckling of the octahedral CoO_6 network, which also is suggested by another author [17]. We also can adopt the Goldschmidt tolerance factor t to qualitatively estimate the structural distortion degree in peroskites with the general formula $RCoO_3$. The following is the expression equation [18]:

$$t = \frac{r_R + r_O}{\sqrt{2}(r_{Co} + r_O)} \quad (1)$$

where r_R, r_{Co} and r_O represent the average ionic radii of the respective cations R, Co and anion oxygen, respectively. Assume that the average ionic radius of Co and O is invariable at room temperature. Therefore, since the ion radius of Sr^{2+} (1.12 Å) is bigger than Y^{3+} (0.89 Å), the tolerance factor t increases with increasing doped Sr content x, leading to the decrease in lattice distortion as x increases [19]. The decrease of distortion may be caused by the partial substitution of Sr for Y, and this also would affect the transport properties of $Y_{1-x}Sr_xCoO_3$ (see following sections), as suggested by Liu et al. [5], Mehta et al. [11], Knížek et al. [12] and Ji-Woong Moon et al. [13].

3.2. Resistivity and its temperature dependences for $Y_{1-x}Sr_xCoO_3$

Fig. 2 plots the dc electrical resistivity ρ as a function of temperature T for $Y_{1-x}Sr_xCoO_3$ ($x=0$, 0.01, 0.05, 0.15, and 0.20) in the temperature range from 20 to 720 K. We note that at low temperatures $YCoO_3$ is a good insulator (6.54×10^3 Ω m at~190 K), which also is reported in a similar system $LaCoO_3$ [20], and with increasing temperature its electrical

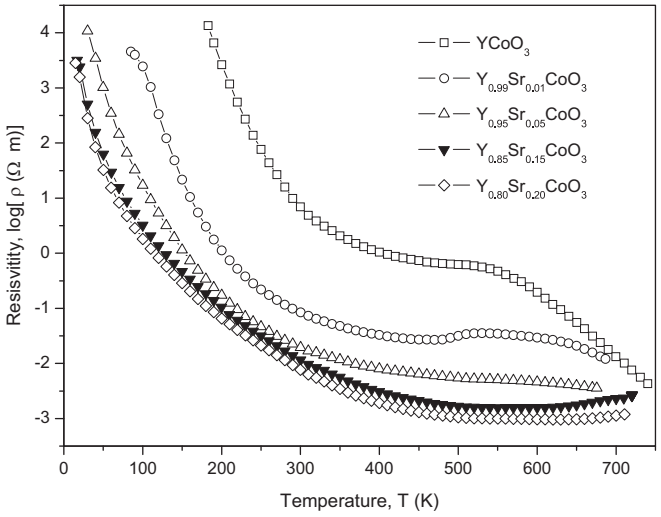

Fig. 2. Electrical resistivity ρ as a function of temperature T for $Y_{1-x}Sr_xCoO_3$ ($x=0$, 0.01, 0.05, 0.15, 0.2).

resistivity rapidly decreases to 1.05 Ω m (at 400 K); in the temperature range 400 K < T < 550 K the resistivity slowly decreases to 0.47 Ω m (at 550 K), where a semiconducting state is suggested [21]; then the resistivity, again, decreases fleetingly (such as from 0.47 Ω m at 550 K down to 7.57×10^{-2} Ω m at 720 K) in temperature range $T > \sim$ 550 K, which is a transition to the higher-temperature metallic state [21]. The data of $YCoO_3$ is basically in agreement with what is reported by Hejtmánek et al. [9,10]. After Sr substitution the electrical resistivity of $Y_{1-x}Sr_xCoO_3$ at the same temperature decreases rapidly with increasing Sr substitution content. For example, the electrical resistivity of $Y_{1-x}Sr_xCoO_3$ at 300 K decreases from 6.92 Ω m for $x=0$ to 8.45×10^{-2} Ω m for $x=0.01$, then it decreases from 1.94×10^{-2} Ω m for $x=0.05$ to 1.15×10^{-2} Ω m for $x=0.15$ and finally arrives at 7.63×10^{-3} Ω m for $x=0.2$. The decrease in electrical resistivity may come from the decrease of distortion degree after Sr substitution (as showed by the increase of t values). The decrease of distortion with increasing Sr content will lead the average Co ($3d$)–O($2p$)–Co($3d$) bond angle to tend to move toward 180°, making the Co($3d$)–O($2p$) orbit overlap to become larger and interactions to become stronger. Further, this decreases the electrical resistivity of $Y_{1-x}Sr_xCoO_3$ as the Sr content of doping increases. This explanation is consistent with what Zaanen et al. [22,23] advocated and the theoretical model [24,25].

In order to more detailedly understand the temperature behavior of electrical resistivity for $Y_{1-x}Sr_xCoO_3$, plots of logarithm of dc electrical resistivity ρ against reciprocal of temperature $10^3/T$ for the compounds $Y_{1-x}Sr_xCoO_3$ are shown in Fig. 3. We note that there are good linear relations between $\ln \rho$ and $1/T$ for the light substituted samples ($x \leq 0.01$). However, for the heavy substituted samples ($x \geq 0.05$) the relation between $\ln \rho$ and $1/T$ deviates seriously from the linear relationship, which results from the change of localization degree with increasing Sr substitution content, as suggested in our investigation for $Y_{1-x}Ca_xCoO_3$ series [4,5]. The

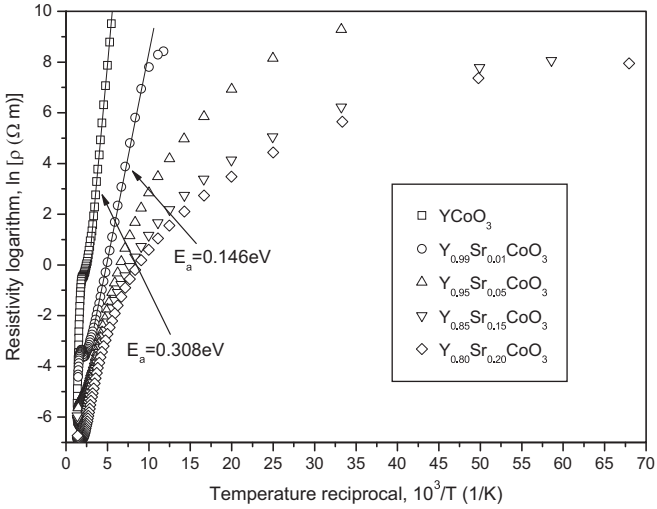

Fig. 3. Electrical resistivity logarithm $\ln \rho$ versus temperature reciprocal $1/T$ for $Y_{1-x}Sr_xCoO_3$ ($x=0$, 0.01, 0.05, 0.15, 0.2).

electrical resistivity data in the linear regions are fitted to Arrhenius relation

$$\rho = \rho_0 \, exp\left(\frac{E_a}{k_B T}\right) \tag{2}$$

where ρ_0 is a constant, E_a is the activation energy for conduction, k_B, the Boltzmann constant, and T, the absolute temperature. By best fit of the experimental data to Eq. (2) for unsubstituted sample $x=0$ and light substituted sample $x=0.01$, we find out that the activation energy of conduction E_a decreases from 0.308 eV for $x=0$ to 0.146 eV for $x=0.01$ with increasing Sr content. This indicates from the p-type semiconductor energy band theory that the Fermi level E_F would shift toward the edge of valence band, which makes the energy difference E_a between valence band edge E_v and Fermi level to become small. This is why the electrical resistivity of $YCoO_3$ has a large decrease after Sr substitution ($x=0.01$).

From the discussion above, we know that the Sr substitution led to the decrease of electrical resistivity in $YCoO_3$, introducing also defects into the lattice in $YCoO_3$. Well then, what is the conduction carrier in $Y_{1-x}Sr_xCoO_3$? The following is a qualitative discussion from the defect chemistry using the dilute solution model and the Kröger–Vink notation for $Y_{1-x}Sr_xCoO_3$. At high oxygen partial pressure and low temperatures the replacement of Y^{3+} by Sr^{2+} is electrically compensated by forming the Co^{4+} cation. The equation is expressed as

$$YCoO_3 \overset{SrO}{\rightarrow} Y^{\times}_{1-x}Sr^{'}_{x}Co^{\times}_{1-x}Co^{\cdot}_{x}O_3 \tag{3}$$

where $Y^{\times}$ ($Sr^{'}$, $Co^{\times}$ and $Co^{\cdot}$) is a Y^{3+} (Sr^{2+}, Co^{3+} and Co^{4+}) cation on a Y^{3+} (Y^{3+}, Co^{3+} and Co^{3+}) lattice position, respectively. The electrical neutrality conduction of Eq. (3) can be written as

$$[Sr^{'}_{Y}] = [Co^{\cdot}_{Co}] \tag{4}$$

At low oxygen partial pressures and/or high temperatures, the following reaction occurs with the formation of oxygen vacancies, assuming the oxygen vacancies are doubly ionized.

$$Y^{\times}_{1-x}Sr^{'}_{x}Co^{\times}_{1-x}Co^{\cdot}_{x}O_3 \Leftrightarrow Y^{\times}_{1-x}Sr^{'}_{x}Co^{\times}_{1-x+2\delta}Co^{\cdot}_{x-2\delta}O_{3-\delta}\left[\delta V^{\cdot\cdot}_{O}\right] + \frac{\delta}{2}O_2 \tag{5}$$

The electroneutrality is the following

$$[Sr^{'}_{Y}] = [Co^{\cdot}_{Co}] + 2[V^{\cdot\cdot}_{O}] \tag{6}$$

As suggested for $LaCoO_3$ [26], above 200 K, its electrical conduction occurs via the transfer of electrons from low-spin cobalt ions Co^{III} ($t_{2g}^6 e_g^0$) to high-spin Co^{3+} ($t_{2g}^4 e_g^2$) giving rise to Co^{IV} and Co^{2+} according to the equation

$$Co^{III} + Co^{3+} \rightarrow Co^{IV} + Co^{2+} \tag{7}$$

This is also expected to take place in $Y_{1-x}Sr_xCoO_3$ series. Therefore, the total expression equation is the following:

$$[Sr^{'}_{Y}] + [Co^{'}_{Co}] = [Co^{\cdot}_{Co}] + 2[V^{\cdot\cdot}_{O}] \tag{8}$$

That is, with increasing Sr content of substitution, the Co^{4+} increases; $[Co^{\cdot}_{Co}]$ and/or $[V^{\cdot\cdot}_{O}]$ can be regarded as the conduction carrier. In literature 5, we discussed that the main contribution to electrical properties of $Y_{1-x}Ca_xCoO_3$ is $[V^{\cdot\cdot}_{O}]$-species, whereas, the contribution of $[V^{\cdot\cdot}_{O}]$ species is negligibly small, indicating that the Co^{4+} exists.

3.3. Thermopower of $Y_{1-x}Sr_xCoO_3$

Fig. 4 presents the temperature dependence of Seebeck coefficient S for $Y_{1-x}Sr_xCoO_3$ ($x=0$, 0.01, 0.05, 0.15, 0.20) in the temperature range from 300 to 780 K. One can note that the Seebeck coefficient values for $Y_{1-x}Sr_xCoO_3$ are all positive in the temperature range of measurement, indicating that the major carriers for conduction are holes. With increasing temperature the Seebeck coefficient of $YCoO_3$ decreases slowly from ~1048 μV/K at 300 K to ~906 μV/K at 550 K; then it rapidly decreases to ~191 μV/K at 760 K. This behavior is in agreement with that of electrical resistivity above, and its temperature behavior here is also basically

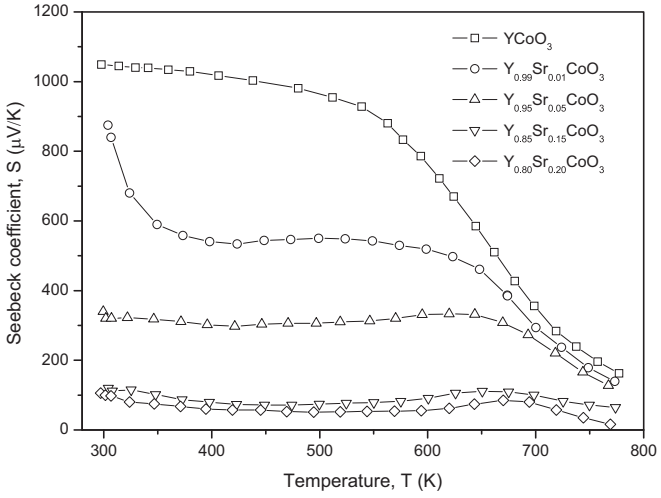

Fig. 4. Temperature dependence of Seebeck coefficient S for $Y_{1-x}Sr_xCoO_3$ ($x=0$, 0.01, 0.05, 0.15, 0.2).

consistent with those from the other authors [9,10,17]. With increasing Sr content of substitution, the temperature dependence of $Y_{1-x}Sr_xCoO_3$ basically has no large change; however, the absolute values of Seebeck coefficient of $Y_{1-x}Sr_xCoO_3$ decrease monotonously. For instance, at 400 K the Seebeck coefficient values decrease from 1020 μV/K for x=0 to 540 μV/K for x=0.01, 301 μV/K for x=0.05, 79 μV/K for x=0.15, 59 μV/K for x=0.2. This monotonous decrease of Seebeck coefficient for $Y_{1-x}Sr_xCoO_3$ in the vicinity of room temperature can be estimated, based on the changes of spin states of Co ions (Co^{3+} and Co^{4+}), by using the temperature-independent Seebeck coefficient described by so-called Heikes formula [27]

$$S = -\frac{k_B}{e} ln\left[\frac{g_3}{g_4}\frac{y}{1-y}\right] \quad (9)$$

where g_3 and g_4 are the number of the degenerated configuration of the Co^{3+} and Co^{4+} states, respectively, y ($=Co^{4+}/Co$) is the concentration of Co^{4+} holes on the Co sites. Applying this formula (9) and the Seebeck coefficient experimental values at 400 K, we obtain the y values 0.0000436, 0.0113, 0.154, 0.706, and 0.752 corresponding to the samples x=0, 0.01, 0.05, 0.15, and 0.20, respectively, assuming that the spin states of Co^{3+} and Co^{4+} at 400 K exist in low spin state $t_{2g}^6$and t_{2g}^5, respectively, that is, g_3=1 and g_4=6. We note that the y values (concentration of Co^{4+}) increase with increasing Sr content of substitution; this is in agreement with the above theory discussion of the Eq. (8). Therefore, the decrease of Seebeck coefficient with increasing Sr content can be well explained based on the framework of Heikes theory.

3.4. Power factor of $Y_{1-x}Sr_xCoO_3$

The temperature dependences of power factor P ($=S^2/\rho$) for the oxides $Y_{1-x}Sr_xCoO_3$ ($0\leq x\leq 0.2$) in the temperature range from 300 to 725 K are shown in Fig. 5. The power factor P was calculated by using the measured electrical resistivity ρ and the Seebeck coefficient S data shown in Figs. 2 and 4, respectively. We note that P of $Y_{1-x}Sr_xCoO_3$ increases remarkably with increasing content of Sr substitution as $x\leq 0.5$. For instance, at 650 K P increases from 5.68×10^{-6} W/m K^2 for x=0 to 9.95×10^{-6} W/m K^2 for x=0.01; then P increases to 2.69×10^{-5} W/m K^2 as x increases to 0.05. This increase of P is mainly due to the larger decrease in the resistivity after Sr substitution. However, as x increases further to 0.2, P decreases to 6.07×10^{-6} W/m K^2, which mainly arises from the large decrease in Seebeck coefficient. The present result indicates that proper Sr substitution for Y in $Y_{1-x}Sr_xCoO_3$ can effectively enhance high-temperature thermoelectric properties of $YCoO_3$ system.

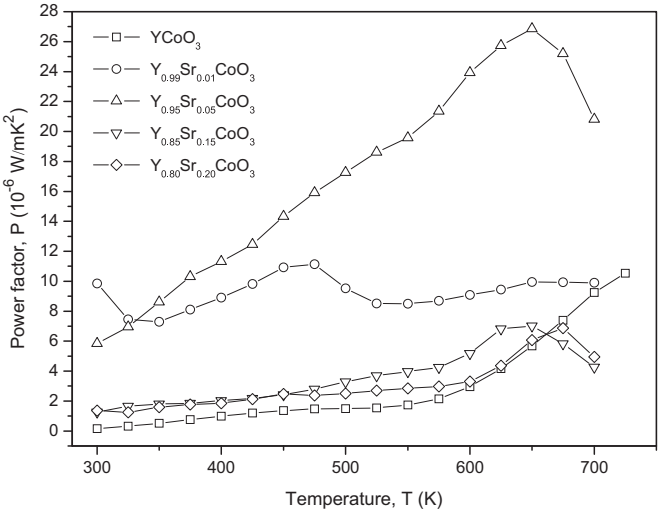

Fig. 5. Temperature dependence of power factor P for $Y_{1-x}Sr_xCoO_3$ (x=0, 0.01, 0.05, 0.15, 0.2).

4. Conclusions

Strontium substituted cobalt oxides $Y_{1-x}Sr_xCoO_3$ ($0\leq x\leq 0.2$) have been prepared by using sol-gel process and its electrical transport and thermoelectric properties have been investigated in the temperature range from 20 to 780 K. The results show that with increasing Sr content of substitution dc electrical resistivity of $Y_{1-x}Sr_xCoO_3$ decreases remarkably, which can come from the contribution of the reduction in lattice distortion. The Seebeck coefficients of $Y_{1-x}Sr_xCoO_3$ are all positive indicating predominant hole-type charge carriers, and they decrease monotonously with increasing Sr content of substitution, which also is explained by the Heikes equation. Moreover, experiments show that the power factor changes non-monotonously with Sr substitution with a maximum value of 2.69×10^{-5} W/m K^2 at 650 K for the sample with x=0.05, which is about five times greater than that for x=0, indicating that appropriate Sr substitution cause effective enhancement in high-temperature thermoelectric properties of $Y_{1-x}Sr_xCoO_3$.

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